

REMARKS

In a supplemental Final Office Action, the Examiner¹ rejected claims 1-6 and 20-23 under 35 U.S.C. § 103(a) over Makasatka (JP 11-271965, "Makasatka") in view of Sato et al. (JP 08-262721, "Sato"). Applicants respectfully traverse the rejections as the cited art does not teach each and every element recited in independent claims 1 and 6, nor is there a motivation to combine the references in the manner alleged by the Examiner.

In order to establish a *prima facie* case of obviousness, three basic criteria must be met. First, the prior art reference (or references when combined) must teach or suggest all the claim elements. Furthermore, "[a]ll words in a claim must be considered in judging the patentability of that claim against the prior art." See M.P.E.P. § 2143.03, 8th Ed., Aug. 2001, p. 2100-126, quoting *In re Wilson*, 165 U.S.P.Q. 494, 496 (C.C.P.A. 1970). Second, there must be some suggestion or motivation, either in the references themselves or in the knowledge generally available to one of ordinary skill in the art, to modify a reference or to combine reference teachings. Third, there must be a reasonable expectation of success. See M.P.E.P. § 2143- § 2143.03.

Claim 1 recites, "[a] pattern formation method for electron beam lithography ... wherein said pattern formation material further comprises first and second dissolution inhibiting groups each of which has a capacity of inhibiting dissolution of said alkali-soluble resin in an alkali solution and loses the capacity upon application of an acid," and further recites that the first and second inhibiting group respectively increases or

¹ The Office Action contains a number of statements reflecting characterizations of the related art and the claims. Regardless of whether any such statement is identified herein, Applicants decline to automatically subscribe to any statement or characterization in the Office Action.

decreases the “sensitivity of said pattern formation material when left to stand in a vacuum after being irradiated with an electron beam.” Applicants respectfully submit that Masataka and Sato et al., taken alone or in combination, do not teach or suggest at least these elements of Applicants’ independent claim 1.

Masataka is characterized by the Examiner as drawn to “a pattern formation method utilizing a positive type chemical amplification resist containing a polymer,” which further comprises “a photoacid generator and a solvent.” (Office Action at 2.) However, the Examiner admits deficiencies of Masataka, and relies on the disclosure of Sato. The Examiner asserts that Sato teaches or suggests a “positive type resist composition having high sensitivity, high resolution and high heat resistance, excellent aging stability, and capable of forming a resist pattern excellent in profile shape,” and that such a resist is “achieved by replacing 10-60 mol% of the hydroxyl group of a poly hydroxyl styrene polymer with tert-butoxycarbonyloxy groups.” (Office Action at 2.) Applicants respectfully disagree and submit that Sato does not teach “an alkali-soluble resin and a photoacid generator,” which “further comprises first and second dissolution inhibiting groups,” as recited in claim 1.

Sato discloses “resin component of the positive resist composition whose solubility into alkaline aqueous solution increases by an action of acid is mixed with substituted polyhydroxy styrenes which include different two specific substituents, respectively, and an organic carboxylic compound is further added thereto,” alleged by the Examiner to form a positive resist composition. See attached partial translation of Sato, paragraph [0008], emphasis added. Sato consistently teaches the requirement of the addition of organic carboxylic compound to form “a resist

pattern,” to obtain excellent cross sectional shape and aging stability. See attached partial translation of Sato, paragraph [0008], [0054], [0060], and [0064]. Therefore, the disclosure of Sato teaches that the addition of a organic carboxylic compound is necessary to form the alleged excellent positive resist, but does not teach benefits of replacing 10-60 mol% of the hydroxyl group of a poly hydroxyl styrene polymer with tert-butoxycarbonyloxy groups, as asserted by the Examiner.

In addition, the Examiner’s allegation that Sato teaches a “positive type resist composition,” which possesses “excellent in aging stability,” and is “capable of forming a resist excellent in profile shape,” at page 2 of the supplemental Office Action is not supported by the cited reference. Applicants note that the alleged excellent aging stability disclosed by Sato is limited to 15 minutes (see attached partial translation of Sato paragraphs [0077], [0079], [0111]), and not 10 hours or longer asserted by the Examiner in the Office Action at page 3. Comparing the Examiner’s stated definition of an excellent positive resist, and the disclosure of Sato, it is unreasonable to equate the so-called excellent aging stability of Sato and the “pattern formation material . . . left to stand in a vacuum,” as recited in claim 1.

Moreover, Sato merely discloses a rectangular *profile* that does not change over time as stated in paragraphs [0066]-[0079] and [0103]-[0112] of the attached machine translation. However, Sato does not disclose the claimed “size of an alkali-soluble portion . . . [that] is substantially held constant independently of a standing time in a vacuum”, as recited in claim 1. (Emphasis added.)

Furthermore, the chemical arts are unpredictable. Even if the Examiner’s alleged compound corresponds to the “pattern formation material,” and Applicants do not agree

that it does, the cited references do not contain any reason to combine them in the alleged combination, and thus the Examiner has not provided the necessary motivation to combine the references.

Applicants note that the Federal Circuit has repeatedly held that it is legal error to find a claimed compound obvious under 35 U.S.C. § 103(a) solely because the prior art discloses a compound that is structurally similar. See e.g., *In re Lalo* 747 F.2d 703 (Fed. Cir. 1984). Instead, the Examiner is required to find that the art not only discloses such a compound but that the art also provides the motivation to make the proposed modifications. *Id.*

Similarly, in *In re Grabiak*, the Federal Circuit reversed the Board's decision and held that Appellants' claimed compounds were not obvious in view of "structurally similar" compounds disclosed in the prior art. 769 F.2d 729 (Fed. Cir. 1985). First, the Court reviewed the case law regarding whether structural similarity is enough to render compounds *prima facie* obvious. In this respect, the Court stated that "[u]pon review of this history, we have concluded that generalization should be avoided insofar as specific chemical structures are alleged to be *prima facie* obvious one from the other." *Id.* at 731.

Instead, "there must be adequate support in the prior art for the . . . change in structure, in order to complete the PTO's *prima facie* case, and shift the burden of going forward to the applicant." *Id.* (emphasis added). After reviewing the references cited by the PTO in support of the determination of obviousness, the court found that the references lacked the necessary motivation to make the ester/thioester modification. Instead, the teachings were merely general in nature. In this respect the court said, "the

Board held that ‘it is not inconceivable to substitute . . . to obtain compounds having the same expected properties.’ We agree that it is not inconceivable. The standard, however, is whether it would have been obvious in terms of section 103.” *Id.* at 733. Thus, the Court reversed the finding of obviousness in view of the Board’s failure to cite any motivation in the art to make the proposed modifications. *Id.*

Even if the Examiner’s alleged compound is similar to that recited in claim 1, the Examiner must also show that such a similarity contributes to a motivation to combine in the asserted manner in the cited references themselves. Since the pattern formation method taught by Masataka, teaches a “polymer,” having an “acid disassociation group of acetal type,” and an “acid generating agent,” (Masataka, Abstract) and Sato teaches the beneficial nature of the addition of an organic carboxylic compound, the Examiner’s alleged compound which results requires an organic carboxylic compound. One of ordinary skill in the arts would not find motivation in the cited references to combine them in a manner asserted by the Examiner to achieve the combination recited in claim 1.

Claim 1 is therefore allowable, as Masataka and Sato, neither alone nor in combination, teach or suggest each and every element recited in claim 1. Moreover, there is not a motivation to combine the cited references in the Examiner’s alleged manner. Further, claim 6, though of different scope, recites similar limitations to claim 1, and is allowable for at least the reasons presented above with respect to claim 1. Claims 2-5 and 20-23 respectively depend from independent claims 1 and 6, and are therefore allowable at least due to their respective dependencies. Accordingly,

Applicants respectfully request the Examiner reconsider and withdraw the rejection to claims 1-6 and 20-23.

In view of the foregoing amendments and remarks, Applicant respectfully requests reconsideration and reexamination of this application and the timely allowance of the pending claims.

Please grant any extensions of time required to enter this response and charge any additional required fees to our deposit account 06-0916.

Respectfully submitted,

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Attachments: Partial Translation of Sato et al. (JP 09-262721)
including paragraphs [0008], [0054], [0060], [0064], [0066]
-[0079], [0103]-[0112]

PARTIAL TRANSLATION OF SATO ET AL.

[0008] The present inventors have devoted themselves to the study for developing a positive chemically amplified resist composition free from the above-described problems. As a result, the present inventors have found that when the resin component of the positive resist composition whose solubility into alkaline aqueous solution increases by an action of acid is mixed with substituted polyhydroxy styrenes which include different two specific substituents, respectively, and an organic carboxylic compound is further added thereto, a positive resist composition with high sensitivity, high resolution, excellent heat resistance and aging stability, and capable of forming a resist pattern excellent in cross sectional shape irrespective of a variety of substrates can be obtained, thereby accomplishing the present invention.

[0054] In the present invention, by adding the organic carboxylic compound, a resist pattern excellent in cross sectional shape can be obtained, and excellent aging stability after light exposure is achieved. In addition, a resist pattern excellent in cross sectional shape can be formed on various substrates.

[0060] The aromatic carboxylic compounds represented by the above general formulae 35 and 36 may be used alone. Alternatively, two or more of them may be mixed together. By blending the aromatic carboxylic compounds, not only a resist pattern excellent in cross sectional shape can be formed, but also good profile shape can be formed irrespective of duration from light exposure to heat treatment as aging stability after light exposure is excellent. Particularly, the aromatic carboxylic compounds represented by the general formula 36 is preferred as a rectangular cross sectional shape can be formed.

[0064] The positive chemically amplified resist composition according to the present invention can further contain miscible additives, for example, commonly used additives such as additional resins for improving a property of resist film, plasticizers, stabilizers, colorants, or surfactants, if desired.

[0066]

[Examples] Although the present invention is detailed described by the following manufacture examples and practical examples, the present invention should not be limited by the examples.

[0067] Manufacture example 1 (synthesis of polyhydroxy styrene in which 8 mol% of hydroxy groups are substituted by tert-butoxycarbonyloxy groups)

120g of polyhydroxy styrene with weight-average molecular weight of 20,000 was dissolved into 680g of N,N-dimethylacetamide. 17.4g of di-tert-butyl-di-carbonate was added to the solution. After stirring to completely dissolve it, spending about 15 minutes, 59g of triethylamine was dropped into the solution while stirring. After the dropping was finished, the solution was kept stirred for about 3 hours. Then, pure water twelve time as much as the obtained solution was added thereto, and the solution was stirred to precipitate a polyhydroxy styrene whose hydroxy groups were substituted by tert-butoxycarbonyloxy groups. The precipitate was washed with pure water, dewatered, and dried, thereby obtaining 125g of polyhydroxy styrene in which 8 mol% of hydroxy groups were substituted by tert-butoxycarbonyloxy groups.

[0068] Manufacture example 2 (synthesis of polyhydroxy styrene in which 35 mol% of hydroxy groups are substituted by tert-butoxycarbonyloxy groups)

The same process as the manufacture example 1 except that quantity of di-tert-butyl-di-carbonate added was changed to 76.5g was carried out to obtain 145g of polyhydroxy styrene in which 35 mol% of hydroxy groups were substituted by tert-butoxycarbonyloxy groups.

[0069] Manufacture example 3 (synthesis of polyhydroxy styrene in which 39 mol% of hydroxy groups are substituted by tert-butoxycarbonyloxy groups)

The same process as the manufacture example 1 except that quantity of di-tert-butyl-di-carbonate added was changed to 85.0g was carried out to obtain 150g of polyhydroxy styrene in which 39 mol% of hydroxy groups were substituted by tert-butoxycarbonyloxy groups.

[0070] Manufacture example 4 (synthesis of polyhydroxy styrene in which 70 mol% of hydroxy groups are substituted by tert-butoxycarbonyloxy groups)

The same process as the manufacture example 1 except that quantity of di-

tert-butyl-di-carbonate added was changed to 153g was carried out to obtain 180g of polyhydroxy styrene in which 70 mol% of hydroxy groups were substituted by tert-butoxycarbonyloxy groups.

[0071] Manufacture example 5 (synthesis of polyhydroxy styrene in which 35 mol% of hydroxy groups are substituted by ethoxyethoxy groups)

120g of polyhydroxy styrene with weight-average molecular weight of 20,000 was dissolved into 680g of N,N-dimethylacetamide. 37.2g of 1-chloro-1-ethoxyethane was added to the solution. After stirring to completely dissolve it, spending about 30 minutes, 78.8g of triethylamine was dropped into the solution while stirring. After the dropping was finished, the solution was kept stirred for about 3 hours. Then, pure water twelve time as much as the obtained solution was added thereto, and the solution was stirred to precipitate 130g of polyhydroxy styrene whose hydroxy groups were substituted by 1-ethoxyethoxy groups.

[0072] Manufacture example 6 (synthesis of polyhydroxy styrene in which 8 mol% of hydroxy groups are substituted by methoxy-n-propyloxy groups)

120g of polyhydroxy styrene with weight-average molecular weight of 20,000 was dissolved into 680g of N,N-dimethylacetamide. 8.6g of 1-chloro-1-methoxypropane was added to the solution. After stirring to completely dissolve it, spending about 30 minutes, 78.8g of triethylamine was dropped into the solution while stirring. After the dropping was finished, the solution was kept stirred for about 3 hours. Then, pure water twelve time as much as the obtained solution was added thereto, and the solution was stirred to precipitate a polyhydroxy styrene whose hydroxy groups were substituted by 1-methoxy-n-propyloxy groups. The precipitate was washed with pure water, dewatered, and dried, thereby obtaining 125g of polyhydroxy styrene in which 8 mol% of hydroxy groups were substituted by 1-methoxy-n-propyloxy groups.

[0073] Manufacture example 7 (synthesis of polyhydroxy styrene in which 39 mol% of hydroxy groups are substituted by methoxy-n-propyloxy groups)

The same process as the manufacture example 6 except that quantity of 1-chloro-1-methoxypropane added was changed to 42.3g was carried out to obtain 130g of polyhydroxy styrene in which 39 mol% of hydroxy groups were substituted by methoxy-n-propyloxy groups.

[0074] Manufacture example 8 (synthesis of polyhydroxy styrene in which 70 mol% of hydroxy groups are substituted by methoxy-n-propyloxy groups)

The same process as the manufacture example 6 except that quantity of 1-chloro-1-methoxypropane added was changed to 75.6g was carried out to obtain 150g of polyhydroxy styrene in which 70 mol% of hydroxy groups were substituted by methoxy-n-propyloxy groups.

[0075] Practical example 1

1.48g of the polyhydroxy styrene (weight-average molecular weight of 20,000) obtained in manufacture example 2 in which 35 mol% of hydroxy groups were substituted by tert-butoxycarbonyloxy groups and 1.48g of the polyhydroxy styrene (weight-average molecular weight of 20,000) obtained in manufacture example 5 in which 35 mol% of hydroxy groups were substituted by ethoxyethoxy groups were dissolved into 16.8g of propylene glycol monomethyl ether acetate. Then, 0.148g of bis(cyclohexylsulfonyl)diazomethane, 0.093g of benzophenone, and 0.0032g of o-hydroxy benzoic acid were further added and dissolved therein. The obtained solution was filtrated by using a membrane filter with a pore diameter of 0.2 μ m to prepare a coating solution for positive resist.

[0076] The coating solution thus prepared was applied to a silicon wafer of 6 inch diameter with use of a spinner and was dried on a hot plate at 90°C for 90 seconds to obtain a resist film of 0.7 μ m thickness. The film was exposed to an excimer laser light through a test chart mask using a reduction projection exposure system NSR-2005EX8A manufactured by Nikon Ltd., heated at 120°C for 90 seconds, subjected to a puddle development for 65 seconds using 2.38 wt% of tetramethylammonium hydroxide aqueous solution, washed with water for 30 seconds, and dried to form a resist pattern. A cross section of the resist pattern thus formed was almost rectangular and satisfactory without affected by standing wave, though it was slightly rounded at its top, and line and space pattern of 0.21 μ m width was formed. In addition, measured was the minimum light exposure at which a surface of the substrate emerged when patterning to a visually recognizable resist pattern having a large area was performed (referred to as the minimum light exposure hereinafter). As a result, the minimum light exposure was 7 mJ/cm². Moreover, heat resistance of a line pattern of 0.5 μ m width thus formed was examined. As a result, its heat

resistance was 130°C.

[0077] A resist pattern was formed by the same method as the above except that standing for 15 minutes was further performed between the light exposure and the heat treatment at 120°C for 90 seconds. As a result, formed was a line and space pattern of 0.21µm width having vertical side surfaces and a satisfactory profile shape.

[0078] Practical example 2

A resist pattern was formed by the same procedures as practical example 1 except that benzophenone was excluded in preparing the coating solution of practical example 1. A cross section of the resist pattern thus formed was almost rectangular and satisfactory, though it was slightly rounded and rippled at its top to such an extent that serious problem did not occur, and line and space pattern of 0.23µm width was formed. In addition, the minimum light exposure was measured. As a result, the minimum light exposure was 8 mJ/cm². Moreover, heat resistance of a line pattern of 0.5µm width thus formed was examined. As a result, its heat resistance was 130°C.

[0079] A resist pattern was formed by the same method as practical example 1 except that standing for 15 minutes was further performed between the light exposure and the heat treatment at 120°C for 90 seconds. As a result, formed was a line and space pattern of 0.23µm width having a satisfactory profile shape.

[0103] Practical example 13

0.9g of the polyhydroxy styrene obtained in manufacture example 3 in which 39 mol% of hydroxy groups were substituted by tert-butoxycarbonyloxy groups and 2.1g of the polyhydroxy styrene obtained in manufacture example 5 in which 35 mol% of hydroxy groups were substituted by ethoxyethoxy groups were dissolved into 16.8g of propylene glycol monomethyl ether acetate. Then, 0.15g of pyrogallol trimesylate and 6.3mg of salicylic acid were further added thereto. The solution obtained by dissolving them was filtrated by using a membrane filter with a pore diameter of 0.2µm to prepare a coating solution for positive resist.

[0104] The coating solution thus prepared was applied to a silicon wafer of 6 inch diameter with use of a spinner and was dried on a hot plate at 90°C for 90 seconds to obtain a resist film of 0.7µm thickness. After writing an image on the film with use of

an electron beam irradiation system HL-8000 manufactured by Hitachi Ltd., the film was heated at 110°C for 90 seconds, subjected to a puddle development for 65 seconds using 2.38 wt% of tetramethylammonium hydroxide aqueous solution, washed with water for 30 seconds, and dried to form a resist pattern. A cross section of the resist pattern thus formed was vertical and satisfactory. At this time, the exposure was measured. As a result, the exposure was 25 $\mu\text{C}/\text{cm}^2$.

[0105] Practical example 14

A coating solution for positive resist was prepared by the same method as practical example 13 except that bis(p-tert-butylphenyl)iodonium trifluoromethane sulfonate was used as an acid generator instead of pyrogallol trimesylate and quantity of the acid generator added was set at 0.15g in practical example 13. Then, a resist pattern was formed by the same procedures as practical example 13. As a result, a contact hole of 0.15 μm diameter was formed, and a cross section of the resist pattern was vertical and satisfactory. At this time, the exposure was 10 $\mu\text{C}/\text{cm}^2$.

[0106] Practical example 15

A coating solution for positive resist was prepared by the same method as practical example 13 except that p-toluenesulfonic acid 2,6-dinitrobenzyl was used as an acid generator instead of pyrogallol trimesylate and quantity of the acid generator added was set at 0.15g in practical example 13.

[0107] Then, a resist pattern was formed by the same procedures as practical example 13. As a result, a contact hole of 0.14 μm diameter was formed, and a cross section of the resist pattern was vertical and satisfactory. At this time, the exposure was 35 $\mu\text{C}/\text{cm}^2$.

[0108] Practical example 16

A coating solution for positive resist was prepared by the same method as practical example 13 except that triphenylsulfonium trifluoromethane sulfonate was used as an acid generator instead of pyrogallol trimesylate and quantity of the acid generator added was set at 0.15g in practical example 13.

[0109] Then, a resist pattern was formed by the same procedures as practical example 13. As a result, a contact hole of 0.14 μm diameter was formed, and a cross section of the resist pattern was vertical and satisfactory. At this time, the exposure was 10 $\mu\text{C}/\text{cm}^2$.

[0110] Comparative example 4

1.48g of the polyhydroxy styrene (weight-average molecular weight of 20,000) obtained in manufacture example 2 in which 35 mol% of hydroxy groups were substituted by tert-butoxycarbonyloxy groups and 1.48g of the polyhydroxy styrene (weight-average molecular weight of 20,000) obtained in manufacture example 5 in which 35 mol% of hydroxy groups were substituted by ethoxyethoxy groups were dissolved into 16.8g of propylene glycol monomethyl ether acetate. Then, 0.148g of bis(cyclohexylsulfonyl)diazomethane and 0.093g of benzophenone were further added and dissolved therein. The obtained solution was filtrated by using a membrane filter with a pore diameter of 0.2 μ m to prepare a coating solution for positive resist.

[0111] A resist pattern was formed by the same method as practical example 1 with use of the coating solution thus prepared. As a result, formed was a line and space pattern of 0.21 μ m width. However, when standing for 15 minutes was further performed between the light exposure and the heat treatment at 120°C for 90 seconds, a pattern with T-shaped configuration was obtained, and the minimum resolution was a line and space pattern of 0.30 μ m width.

[0112]

[Effect of the Invention] The positive resist composition according to the present invention is the chemically amplified one with high sensitivity, high resolution, excellent heat resistance and aging stability, capable of forming a resist pattern excellent in profile shape irrespective of substrate, excellent in response to radiation, in particular ultraviolet radiation, far infrared radiation, excimer laser, X-ray, and electron beam, and particularly effective to fine processing in VLSI manufacturing process.